

LCA Methodology

Development of Damage Function of Acidification for Terrestrial Ecosystems Based on the Effect of Aluminum Toxicity on Net Primary Production

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Abstract

Background. Acidification is one of the important impact categories for life cycle impact assessment. Although its characterization has progressed during this decade through the employment of midpoint approaches, only limited studies of endpoint approaches have been performed.

Objective. This study aimed at developing damage function of acidification for terrestrial ecosystems in Japan. Damage function expresses a quantitative relationship between the inventory and endpoint damage.

Methods. The geographical boundary was limited in Japan both for emission and impact. In this study, sulfur dioxide (SO₂), nitrogen monoxide (NO), nitrogen dioxide (NO₂) (NO and NO₂ collectively mean NO_x), hydrogen chloride (HCl), and ammonia (NH₃) were considered as major causative substances of acidification. Net primary production (NPP) of existing vegetation was adopted as an impact indicator of terrestrial ecosystems. The aluminum toxicity was adopted as the major factor of effect on terrestrial ecosystems due to acidification. The leachate concentration of monomeric inorganic aluminum ions was selected to express the plant toxicity of aluminum.

Results and Discussion. The results of damage function gave utilizable factors both for a midpoint approach and an endpoint approach; Atmospheric Deposition Factor (ADF) and Damage Factor (DF) applicable to the former and the latter, respectively. The ADF indicates an increase of H⁺ deposition per unit area to an additional emission of causative substance. The additional emission corresponds to some alternatives in industry, not the baseline emission. The DF indicates the total NPP damage in all of Japan due to the additional emission of causative substances. The derived NPP damage is on the order of one millionth of the NPP itself. HCl and NH₃ showed larger ADFs and DFs than that of SO₂ and NO_x. The reason was ascribed to the relatively large source-receptor relationships (SRR) of HCl and NH₃. However, since the method applied to determine the SRR of HCl and NH₃ has larger uncertainties than that of SO₂ and NO_x, attention is needed to handle the difference.

Conclusion. The damage function easily defines the concrete NPP damage due to an additional emission. The impact indica-

tor, NPP, also has an advantage in its mass unit that is directly summable through the entire impact categories. Expansion of endpoints, such as in aquatic ecosystems, material degradation, human health, and biodiversity aspects of terrestrial ecosystems, is an important subject for future work. Further, uncertain analyses for major parameters will provide helpful information on the reliability of damage function.

Abbreviations: ADF-Atmospheric Deposition Factor; AP-Acidification Potential; CI-Category Indicator; DF-Damage Factor; DI-Damage Indicator; EF-Effect Factor; LCA-Life Cycle Assessment; LCIA-Life Cycle Impact Assessment; LIME-Life-cycle Impact assessment Method based on Endpoint modeling; NDVI-Normalized Difference Vegetation Index; NNR-Non-Neutralization Ratio; NPP-Net Primary Productivity; NSS-Non-Sea Salt; SRR-Source-Receptor Relationship

Keywords: Acidification; acidifying substance; aluminum toxicity; causative substance; damage function; Japan; life cycle impact assessment; net primary productivity; terrestrial ecosystems

Introduction

Acidification has been involved in many LCA studies as an impact category or an impact pathway, which is one of the important environmental issues. Acidification is caused by atmospheric deposition of acidifying substances such as sulfuric acid (H₂SO₄) and nitric acid (HNO₃), mainly generated from oxidized sulfur and nitrogen which is largely contributed from anthropogenic emitters.

Early LCA studies based on midpoint approach employed a characterization factor, termed acidification potential (AP) as originally developed by Heijungs et al. (1992), to evaluate emissions of causative substances of acidification. AP only gives relative information on emission impact of causative substances on the basis of SO₂ equivalent. Although several later studies (e.g. Potting et al. 1998, Huijbregts 1999) improved the circumstances considering regional sensitivity of ecosystems and atmospheric fate of causative substances, these were still confined to an equivalent load without information on concrete damage caused by acidification.

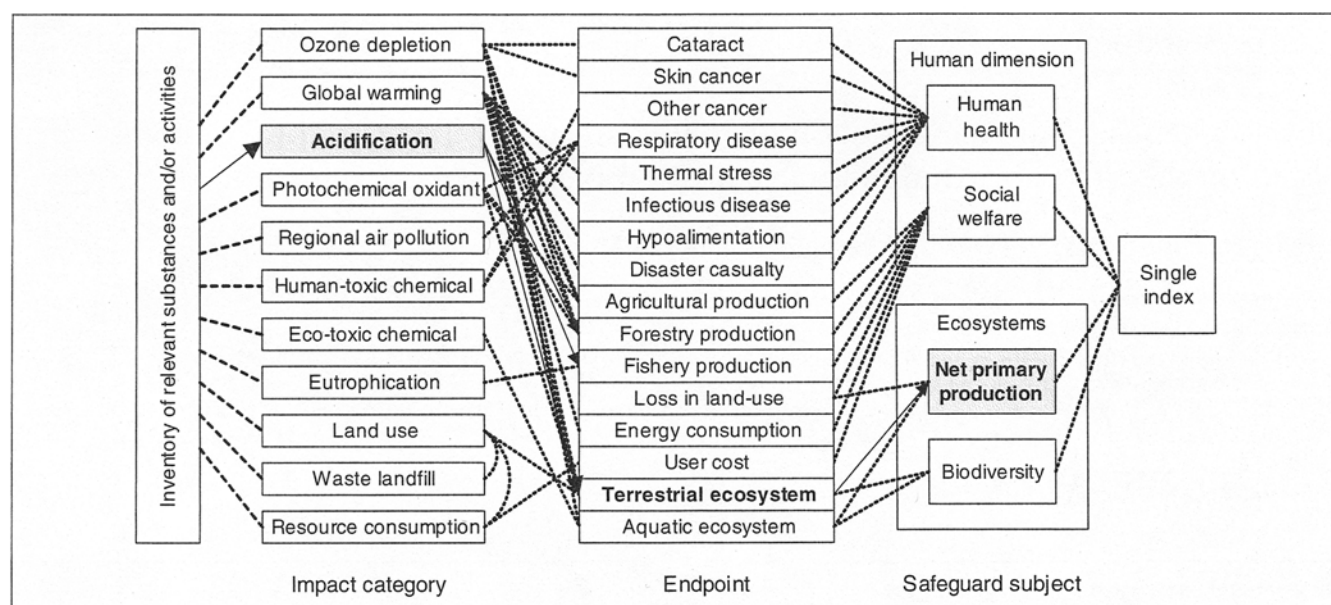


Fig. 1: LCIA framework of LIME

Table 1: Entreatment of 'acidification': in several LCIA frameworks

LCIA framework	Causative substance	Target endpoint relevant to terrestrial ecosystems	Damage indicator	Target region
Eco-indicator 99 ^a	SO _x , NO _x , NH ₃	Species extinction (vascular plants)	PDF (species)	Europe
EPS Version 2000 ^b	SO _x , NO _x , H ₂ S, HF, HCl, NH ₃	Crop growth Wood growth Soil acidification (base cation) Species extinction (life)	Mass (kg) Mass (kg) Capacity (eq) NEX (-)	World
ExternE ^c	Total acidity (from SO ₂ , NO _x)	Agriculture Forest and natural ecosystems	WTP (Euro) WTP (Euro)	Europe
LIME ^d	SO ₂ , NO _x , NH ₃ , HCl	Primary production of vegetation	NPP (kg)	Japan

^aEco-indicator 99 (Goedkoop and Spriensma 2000) includes eutrophication aspects.

^bEPS Version 2000 (Steen 1999) originally deals with the endpoints in Table 1 as impact categories. Therefore, these include all relevant pathways, not limited to acidification.

^cExternE (Holland et al. 1999) includes direct effects of air pollutants such as ozone, SO₂ and NO_x.

^dLIME involves this study, and it deals with regional air pollution as another category.

Given the circumstances, several LCIA frameworks tackled the acidification problem applying endpoint approaches, e.g. Eco-indicator 99 (Goedkoop and Spriensma 2000), EPS (Steen 1999), and ExternE (Holland et al. 1999). In addition, LIME was developed in Japan as the result of the LCA National Project of Japan funded by METI/NEDO (Inaba and Itsuo 2002). LIME is a comprehensive LCIA methodology with endpoint approaches, which also involved acidification in its framework (Fig. 1). Table 1 summarizes the features of each framework narrowed down to category endpoints of acidification regarding terrestrial ecosystems. It shows that the handling of causative substances, target endpoints, and damage indicators are largely different among the frameworks.

This study, forming a part of LIME, focused on the impact on terrestrial ecosystems with priority among various endpoints regarding acidification. A reason was that terrestrial acidification tends to precede aquatic acidification, namely inland water is acidified after the depletion of acid neutralization capacity of its watershed. Moreover, since the major part of land is covered by vegetation, acidification has a potential to induce

remarkable damage to terrestrial ecosystems, i.e. it is a primary producer of ecosystems. As liming activities (e.g. Henrikson et al. 1995; Meiwes 1995) indicate, much work and time are necessary for recovering from acidified conditions. Hence, terrestrial ecosystems are likely to suffer the largest impact due to acidification among various endpoints.

What is a suitable indicator for evaluating acidification impact on terrestrial ecosystems? LIME selected two safeguard subjects regarding ecosystems, i.e. primary production and biodiversity (see Fig. 1). Although both primary production and biodiversity are important safeguard subjects, biodiversity aspects were excluded from this study. Biodiversity impact induced by acidification, however, was difficult to quantify due to the lack of information regarding inherent tolerance against acid for all relevant species and the risk of extinction due to acid stress. In this study, therefore, we only aimed at quantifying the effects on primary production. LIME adopted NPP, a rate of primary production utilizable in food webs, as an impact indicator for terrestrial ecosystems in relation to primary production. NPP is defined as

the difference between gross photosynthesized energy and respiratory-consumed energy by plants; it is expressed by mass as dry matter per unit area and time (e.g. $\text{kg ha}^{-1} \text{yr}^{-1}$). It is considered that NPP is adequate as an impact indicator of terrestrial ecosystems, since NPP stands not only for the fundamental energy supply in food webs, but also for the indispensable oxygen supply in the biosphere. Meanwhile, the existing biomass of a primary producer, as a candidate for an indicator, denotes a stock energy only at a specific time point. Accordingly, existing biomass is inadequate to express the sustainability of terrestrial ecosystems. Although an LCIA study for land use (Lindeijer et al. 1998) employs NPP as an impact indicator, no similar study was found in the field of acidification.

The purpose of this study is to develop the damage function of acidification for terrestrial ecosystems; damage function is defined as quantitative relationship between emission of causative substance and endpoint damage. The prototype of our study was shown in Hayashi et al. (2000).

1 Methods

1.1 Acidifying substance and its causative substance

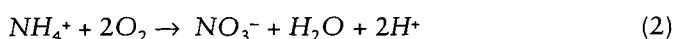
Major acidifying substances in the atmosphere originating from artificially emitted air pollutants are H_2SO_4 and HNO_3 . However, the contribution of HCl, i.e. non-seasalt chloride (NSS-Cl), to dry deposition was observed in Japan as well (Takahashi et al. 2001). Furthermore, NH_3 has a potential of univalent acid in charge balance corresponding to nitrification in soil, though NH_3 is a base in the atmosphere.

Main causative substances of these acidifying substances are SO_2 , NO_x (NO and NO_2), HCl, and NH_3 . A load of acid is expressed by a load of proton (H^+), as determined by the charge balance between total anion and total cation, excluding H^+ on the basis of the Arrhenius acid-base theory expressed with major ion species in the atmosphere by,

$$[\text{H}^+]_{\text{act}}^0 = [\text{SO}_4^{2-}] + [\text{NO}_3^-] + [\text{Cl}^-] - [\text{NH}_4^+] - [\text{Na}^+] - [\text{K}^+] - [\text{Mg}^{2+}] - [\text{Ca}^{2+}] \quad (1)$$

where each term has a unit of charge equivalent concentration (e.g. eq L^{-1}), $[\text{H}^+]_{\text{act}}^0$ is actual acidity without nitrification, and other terms correspond to each ion species, following the sequence in the right side of Eq. 1, sulfate, nitrate, chloride, ammonium, sodium, potassium, magnesium, and calcium.

As mentioned above, nitrification of deposited NH_4^+ acts as an acid, with 2 mol of H^+ per mol of NH_4^+ ,



Inserting Eq. 2 into Eq. 1, actual acidity with complete nitrification after deposition, $[\text{H}^+]_{\text{act}}$, is expressed by,

$$[\text{H}^+]_{\text{act}} = [\text{SO}_4^{2-}] + [\text{NO}_3^-] + [\text{Cl}^-] + [\text{NH}_4^+] - [\text{Na}^+] - [\text{K}^+] - [\text{Mg}^{2+}] - [\text{Ca}^{2+}] \quad (3)$$

In the context of acidification, atmospheric deposition loads of SO_4^{2-} , NO_3^- , Cl^- , and NH_4^+ contribute as sources of potential acidity. Therefore, these ionic species were considered to be potential acidifying substances in this study. Furthermore, these acidifying substances were considered as only the source of acidity in this study. And other aspects like nitrogen as a nutrient were excluded from consideration. In addition, direct effects of air pollutants on terrestrial ecosystems such as oxidants and SO_2 itself were excluded from the impact category of acidification. As shown in Fig. 1, these effects were considered in the impact category of photochemical oxidant and regional air pollution, respectively.

1.2 Cause of damage

There are many schools of thought on causes of forest decline (Hinrichsen 1986). In related to indirect effects caused by acidification, relevant causes are general-multiple stresses, aluminum toxicity, magnesium deficiency, excess nutrient such as nitrogen, heavy metal dissolution, and growth-altering organic chemicals. Among these various causes, the plant toxicity of inorganic aluminum ion, in particular Al^{3+} (Jenny 1961), was selected as the primal factor of acidification impact on terrestrial ecosystems. It partly has to be ascribed to data availability in order to quantitatively relate causes and effects, though the aluminum toxicity is perhaps a serious factor. To express the aluminum toxicity, Al^{3+} concentration in soil solution was adopted.

1.3 Outline of damage function derivation and definition of factors

Fig. 2 shows a schematic diagram of the damage function of acidification. The geographical boundary was limited to Japan, namely both emission and impact only in Japan were considered. The damage function of acidification for terrestrial ecosystems consists of two factors. One is termed Atmospheric Deposition Factor (ADF) and reveals an average

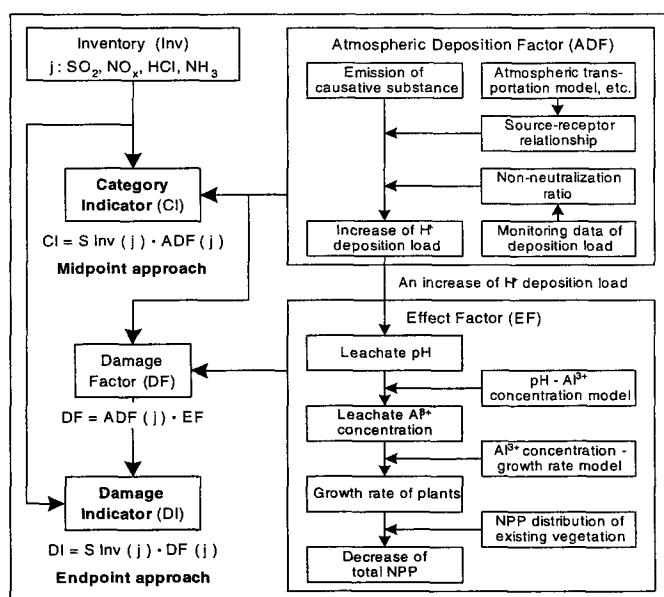


Fig. 2: Schematic diagram of damage function of acidification for terrestrial ecosystems

increase of H^+ load per unit area in Japan due to additional emission of causative substance. The other is termed Damage Factor (DF) which gives a total NPP decrease in all of Japan due to the additional emission of causative substances.

ADF was determined using the fate of an emitted causative substance expressed as the product of Source-Receptor Relationship (SRR) by Non-Neutralization Ratio (NRR) of acid in the atmosphere. SRR denotes the mean deposition fraction of an acidifying substance in Japan due to the emission of its causative substance in Japan. ADF is also utilizable for a mid-point approach; the summation of the products of ADF for each causative substance by its emission amount, namely, stands for the Category Indicator (CI) of acidification.

Meanwhile, DF was given by the product of ADF by Effect Factor (EF) that gives a total NPP decrease in all of Japan due to unit increase of atmospheric deposition load of H^+ per unit area, namely per ADF. EF was defined as the slope of the relationship between the increase of H^+ deposition load per unit area and total NPP decrease in the whole of Japan. The relationship was derived from quantitatively connecting the following processes; increase of H^+ load \rightarrow decrease of leachate pH \rightarrow increase of leachate Al^{3+} concentration \rightarrow decrease of plant growth rate \rightarrow NPP damage. DF is utilizable for an endpoint approach, namely the summation of the products of DF for each causative substance by its emission inventory provides the Damage Indicator (DI) of acidification. DI denotes the concrete damage due to additional emissions of causative substances.

1.4 Calculation of atmospheric deposition factor

1.4.1 Source-receptor relationship of acidifying substance

a. SO_2 and NO_x

In 1991, SRRs of SO_2 and NO_x were derived from the estimation results of regional SRRs for 6 regions of Japan as the annual average values in Japan (Nakaminami 2000). These regional SRRs were calculated by modifying the Eulerian model for atmospheric transportation (Ikeda and Higashino 1997). The values of SRRs were estimated to be 0.136 and 0.140 for SO_2 and NO_x , respectively.

b. HCl

All of the NSS-Cl⁻ in the atmosphere was assumed to be HCl in this study. A value of SRR of HCl in Japan was calculated using existing data on emission and deposition from different sources, since few studies on the SRR of HCl were available.

Waste incineration and fossil fuel combustion were considered as HCl sources, whereas the leakage from chemical industry was neglected. The amount of generated HCl at incineration was calculated by multiplying the amount of incinerated waste (SBJ 1998, 1999, 2000) by the average content of volatile chlorine in waste (NCUC-JWRF 1999) for each year from 1993 to 1997. Then, the amount of atmospheric emission of incineration-origin HCl was determined using the efficiency of flue-gas treatment facilities (Shigaki 1998). As a result, the annual mean of HCl emission from waste incineration was estimated to be 4.20 Gg-Cl yr⁻¹. Next, the amount of generated HCl at combustion was calculated by multiplying the amount of oil and coal consumptions

converted from the primary energy consumption of oil and coal (ANRE 1999) with the average Cl content in oil and coal (Okita 1982) for each year from 1993 to 1997. Then, the amount of atmospheric emission of combustion-origin HCl was determined assuming the same removal efficiency of incineration-origin HCl. The annual means of HCl emission from oil and coal combustion were estimated to be 137 and 3.16 Gg-Cl yr⁻¹, respectively. Hence, the annual mean of the total HCl emission was estimated to be 145 Gg-Cl yr⁻¹.

On the other hand, atmospheric deposition consists of wet and dry deposition. As the sum of wet and dry deposition, the annual deposition of NSS-Cl at the observatories of the Nationwide Survey of Acid Deposition was calculated for each year from 1993 to 1997 using the observation data of Cl⁻ deposition (ADORC 2000) and the composition of major ion species in seawater (JWA 1990); the calculation was based on the assumption that all deposited Na⁺ was of seasalt origin. NSS-Cl deposition was then calculated by subtracting the production of Na⁺ deposition by the ratio of Cl⁻ to Na⁺ in seawater from Cl⁻ deposition. These values, indicating deposition at each observatory, were averaged for urbanized and non-urbanized sites, and the total deposition in all of Japan was then calculated by summing the product of the averaged deposition by the area of each land classification. The annual mean of the total deposition of NSS-Cl was estimated to be 57.0 Gg-Cl yr⁻¹.

The SRR of HCl, derived by dividing the total deposition by the total emission, was 0.404.

c. NH_3

A value of SRR of NH_3 was calculated using existing data in this study similar to that of HCl.

The estimated value of annual NH_3 emission in Japan in 1994 was 460 Gg-N yr⁻¹ (Kannari et al. 2001), including 5% originating from natural sources.

The annual mean of wet deposition of NH_4^+ in Japan from 1987 to 1989 was 336 Gg-N yr⁻¹ (Fujita et al. 2000). This estimation was derived by using a grid system and corresponded to the area of 722,000 km², excluding the area of the Nansei Islands, but including sea areas in each grid. Therefore, the value was simply separated for land using the land area in the same range, 370,000 km², so that 172 Gg-N yr⁻¹ was then obtained. On the other hand, the annual mean of dry deposition of ammoniacal nitrogen at the observatories of the Nationwide Survey of Acid Deposition in 1994 was calculated using the observation data (ADORC 2000). Then, the dry deposition in the whole of Japan was calculated in the same way for HCl. The annual mean of dry deposition of ammoniacal nitrogen was estimated to be 13.0 Gg-N yr⁻¹.

The SRR of NH_3 derived from dividing the sum of wet and dry deposition by the total emission was 0.432.

1.4.2 Non-neutralization ratio of acid

Acidifying substances in the atmosphere are neutralized by coexisting basic substances. Here, it was assumed that acidifying substances which originated from additionally emitted substances were also neutralized as well as in the present

condition. Thus, NNR, defined as the ratio of actual acidity with nitrification to potential acidity in the atmosphere, was calculated as follows using the total deposition of H^+ , NH_4^+ , $NSS-Cl$, NO_3^- , and $NSS-SO_4^{2-}$ at the observatories of the Nationwide Survey of Acid Deposition (ADORC 20000):

$$NNR = (H_{dep} + 2 \cdot NH_{4dep}) / (nssCl_{dep} + NO_{3dep} + nssSO_{4dep} + NH_{4dep}) \quad (4)$$

where each term on the right side denotes the deposition of corresponding ionic species, following the sequence in Eq. 4, proton, ammonium, non-seasalt chloride, nitrate, and non-seasalt sulfate. Each term in the right side has a unit of equivalent load (e.g. $meq\ m^{-2}\ yr^{-1}$). Regarding the numerator, the reason why $2\ NH_{4dep}$ was added to H_{dep} is that this H_{dep} was derived from the observed pH at which NH_4^+ took a role as a base. Therefore, to determine actual acidity with nitrification, the generation of $2\ H^+$ per NH_4^+ must be considered. In addition, NNR is principally related not to each ionic species directly, but to the acidity produced resulting from the total charge balance.

NNR at each observatory was calculated for each year from 1993 to 1997 and they were averaged for urbanized and non-urbanized sites. The weighted mean of NNR was determined using the respective area of urbanized and non-urbanized regions as the weight. Subsequently, the annual weighted mean of NNR from 1993 to 1997 was estimated to be 0.769.

1.5 Calculation of effect factor

1.5.1 Relationship between acidic deposition and soil acidification

It was assumed that an increased H^+ load due to an additional emission of acidifying substance completely contributes to soil acidification, namely decreasing the value of soil pH. Based on the assumption, an increase of H^+ concentration in leachate to an increased H^+ load was estimated; leachate is equal to vertically leaching water from soil. Leachate flux being used to determine an increase of H^+ concentration was simply expressed by,

$$Q = Pr - ET \quad (5)$$

where Q , Pr , and ET denote annual leachate flux, annual precipitation, and annual evapotranspiration, respectively. Each term has a unit of $mm\ yr^{-1}$.

Q was calculated for each grid, termed 10 km grid, with the size of 5.0' for latitude and 7.5' for longitude. Pr was derived from the gridded dataset of meteorology as the normal values from 1961 to 1990 (MAJ 1996). The original dataset was also composed as a grid system with a ten-fold higher resolution than that of the 10 km grid, and the arithmetic mean of Pr for each 10 km grid was calculated. On the other hand, ET was determined by applying the Thornthwaite method (Thornthwaite 1948), an empirical model as a function of monthly mean temperature and latitude. The monthly mean temperature in each 10 km grid was calculated, as well as the Pr employing the same dataset of Pr (MAJ 1996).

An increase of H^+ concentration was determined by dividing an increase of H^+ deposition by Q ,

$$[H^+]_{inc} = (H_{add-dep} / Q) \cdot 10^{-6} \quad (6)$$

where $[H^+]_{inc}$ and $H_{add-dep}$ denote an increase in H^+ concentration ($eq\ L^{-1}$) and increase in H^+ deposition ($eq\ km^{-2}\ yr^{-1}$), respectively.

Next, the representative soil type in each 10 km grid was determined as the most dominant soil type in the grid with the soil map of Japan at a scale of 1:1,000,000 (CSCN-GJP 1990). The value of soil pH for each representative soil type was given as the median value of the corresponding soil type in the dataset of soil pH (Batjes 1995), data which was used to compile the soil unit of FAO-UNESCO Soil Map of the World (FAO-UNESCO 1990).

Soil pH was assumed to be equal to leachate pH, and a value of leachate pH corresponding to an increase of H^+ concentration was then expressed by,

$$pH_1(lea) = -\log(10^{-pH_0(lea)} + [H^+]_{inc}) \quad (7)$$

where $pH_1(lea)$ and $pH_0(lea)$ denote leachate pH with an increase of H^+ concentration and initial leachate pH, respectively.

1.5.2 Relationship between soil acidification and Al^{3+} concentration

Although leachate pH affects leachate Al^{3+} concentration, its quantitative relationship differs among soils. In this study, approximate models for each representative soil type were derived from existing data (So *et al.* 1999; Shioiri 1934; Kitagawa 1966; Yamaya 1968; Umemura 1968; NFRI 1968; Maruyama 1995), which expresses the relationship between leachate pH and leachate Al^{3+} concentration. The derived models are shown in Table 2.

Table 2: Relationship between leachate pH and Al^{3+} concentration derived for representative soil types in Japan

Applied soil type in this study	Exponential models for leachate pH – leachate Al^{3+} concentration ^a	Soil or mineral type in original data
Podzols, Podzolic soils ^b	$[Al^{3+}] = 9.98E+3 \times \exp(-0.585\ pH(lea))$	Podzols
Brown forest soils, Yellow-brown forest soils ^b	$[Al^{3+}] = 9.13E+4 \times \exp(-2.19\ pH(lea))$	Brown forest soils
Red-yellow soils ^b	$[Al^{3+}] = 2.30E+6 \times \exp(-2.65\ pH(lea))$	Red-yellow soils
Andosols ^b	$[Al^{3+}] = 5.33E+8 \times \exp(-3.69\ pH(lea))$	Andosols
Gley soils, Lowland soils, Lithosols ^b	$[Al^{3+}] = 6.78E+2 \times \exp(-1.25\ pH(lea))$	Crystalline clay minerals ^c

^a $pH(lea)$: leachate pH; $[Al^{3+}]$: leachate Al^{3+} concentration ($\mu mol\ L^{-1}$).

^b These models were derived from existing data (So *et al.* 1999, Shioiri 1934, Kitagawa 1966, Yamaya 1968, Umemura 1968, NFRI 1968, Maruyama 1995).

^c The approximate equation of crystalline clay minerals was applied to gley soils, lowland soils, and lithosols, since no information on pH – Al^{3+} relationships was obtained for these soil types.

1.5.3 Relationship between Al^{3+} concentration and plant growth rate

An experimental result with *Pinus densiflora* (Japanese red pine) (Lee et al. 1997) was applied to express the relationship between leachate Al^{3+} concentration and growth rate of trees as dry matter. The derived model is as follows:

$$GR = 8.00 \cdot 10^{-6} \cdot [Al^{3+}]^2 - 0.0314 \cdot [Al^{3+}] + 100 \quad ([Al^{3+}] < 2000) \quad (8)$$

where GR and $[Al^{3+}]$ denote relative growth rate of trees to control (%) and leachate Al^{3+} concentration ($\mu\text{mol L}^{-1}$), respectively.

1.5.4 NPP of existing vegetation and its damage

The mean NPP ($\text{kg ha}^{-1} \text{yr}^{-1}$) of each 10 km grid was calculated using three map sources for existing vegetation, land use, and NDVI, and using a chart of NPP values for representative vegetation types. NDVI is an index of relative photosynthesis activity.

A grid system with $4.5'' \times 4.5''$ resolution, termed 100 m grid, was overlaid on a digital map of existing vegetation, the Dataset for GIS on Natural Environment (EAJ 1999), vector formatted, and the legend of the most dominant vegetation in each grid was then extracted as the representative vegetation of the grid. The original legends of existing vegetation of the map, approximately 900 legends, were integrated and simplified into 30 legends on the chart of NPP values (Iwaki 1981), and these NPP values were then allocated to each 100 m grid corresponding to its legend. The legend of each 100 m grid, however, was replaced by 'others', indicating highly artificial land, when the central point of the 100 m grid corresponded to the legend of a 'building site' or 'arterial road' of another digital map, the Detailed Gridded Map of Land Use in Japan with resolution of $3''$ for latitude and $4.5''$ for longitude (MLIT 2002).

Furthermore, a digital map of NDVI, the Gridded NDVI Dataset with $36'' \times 36''$ resolution (CGER 2000), was overlaid on the produced NPP map of existing vegetation, and then the NDVI value at the central point of each 100 m grid was extracted. Subsequently, these extracted values were averaged for each of 30 legends. The ratio of the NDVI value at the central point of each 100 m grid to the averaged NDVI value corresponding to the legend at the point was applied to correct the fixed NPP value in the chart of Iwaki (1981). In addition, an NPP value of each 100 m grid was only calculated if the grid had all properties on existing vegetation, land use, and NDVI.

Finally, the mean NPP ($\text{kg ha}^{-1} \text{yr}^{-1}$) of each 10 km grid was determined by averaging NPP values for all the 100 m grids in the 10 km grid; here, the weight of 0.5 was applied to 100 m grids which straddled two 10 km grids.

NPP damage in each 10 km grid was calculated as follows:

$$L\text{-NPP}_{act} = NPP_{act} \cdot A \cdot (GR_0 - GR_1) / GR_0 \quad (9)$$

where $L\text{-NPP}_{act}$, NPP_{act} , A , GR_0 , and GR_1 denote NPP damage in 10 km grid (kg yr^{-1}), average NPP of existing vegetation in a 10 km grid ($\text{kg ha}^{-1} \text{yr}^{-1}$), land area of 10 km grid (ha), growth rate at initial condition (%), and growth rate affected by additional H^+ load (%), respectively.

The total NPP damage in the whole of Japan, $L\text{-NPP}_{act}(tot)$, was expressed by,

$$L\text{-NPP}_{act}(tot) = \sum_i^n L\text{-NPP}_{act}(i) \quad (10)$$

where $L\text{-NPP}_{act}(i)$ denotes NPP damage of i -th 10 km grid in Japan.

2 Results

2.1 Atmospheric deposition factor

Due to an additional emission of its causative substance, a total deposition of acidifying substance in all of Japan was obtained by multiplying the additional emission of the substance by its SRR. A mean deposition load per unit area was derived by dividing the total amount by the land area of Japan. However, in the context of acidification, an additional emission amount of a causative substance was necessary to be converted into H^+ load. ADF ($\text{eq km}^{-2} \text{yr}^{-1} \text{kg}^{-1}$), which indicates the mean increase of H^+ deposition ($\text{eq km}^{-2} \text{yr}^{-1}$) in Japan by 1 kg yr^{-1} emission of causative substance, j , was expressed by,

$$ADF(j) = 1 \cdot 10^3 \cdot SRR(j) \cdot MW(j)^{-1} \cdot VA(j) \cdot LA^{-1} \cdot NNR \quad (11)$$

where MW and VA denote molecular weight (g mol^{-1}) and valence of acid of chemical species j , respectively. LA denotes the land area of Japan (km^2). The concrete data to determine ADF are shown in Table 3, and the determined values of ADF are shown in Table 4. ADF is directly applicable to a midpoint approach.

Table 3: Input data for determining Atmospheric Deposition Factors (ADFs)^a

Causative Substance (j)	Source-receptor relationship (SRR)	Molecular weight (MW)	Valence of acid (VA)	Land area of Japan (LA)	Non-neutralization ratio (NNR)
SO ₂	0.136	64.1	2	372,798 km ² (1995)	0.769 (1993–1997) Mean value
NO _x	0.140	30.0	1		
		46.0	1		
HCl	0.404	36.5	1		
NH ₃	0.432	17.0	1		

^a $ADF(j) [\text{eq km}^{-2} \text{yr}^{-1} \text{kg}^{-1}] = 1 \times 10^3 \times SRR(j) \times MW(j)^{-1} \times VA(j) \times LA^{-1} \times NNR$

2.2 Effect factor

Connecting from Eq. 6 to Eq. 10 and inserting the following values of $H_{add-dep}$, 0, 1, 5, 10, 15, 20, 25, 50, 75, 100, 150, and 200 $\text{eq km}^{-2} \text{yr}^{-1}$ into Eq. 6, $L\text{-NPP}_{act}(tot)$ corresponding to each $H_{add-dep}$ was obtained. Plotting these data on the $H_{add-dep}$ - $L\text{-NPP}_{act}(tot)$ coordinate system (Fig. 3), its slope, 36859, was then adopted as EF, which indicates the total annual NPP damage in all of Japan corresponding to the 1 $\text{eq km}^{-2} \text{yr}^{-1}$ increase of mean H^+ load in Japan.

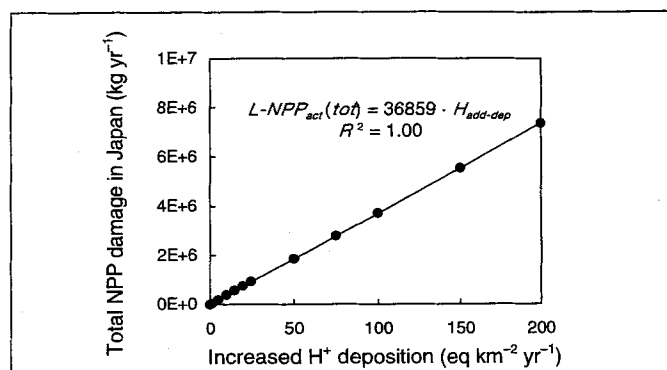


Fig. 3: Relationship between increased proton deposition per unit area and total NPP damage in the whole of Japan

2.3 Damage factor

DF for each causative substance is shown in Table 4, which was obtained by multiplying ADF for each causative substance by EF. DF is directly applicable to an endpoint approach.

Table 4: Damage function of acidification for terrestrial ecosystems

Type 1: Emission unit is in kg of chemical species yr^{-1}

Causative substance		Emission unit	Midpoint approach		Endpoint approach
			ADF ^a	AP ^b	DF ^c
SO ₂		kg-SO ₂ yr^{-1}	8.78 e-6	1.00	0.323
NO _x	NO	kg-NO yr^{-1}	9.61 e-6	1.09	0.354
	NO ₂	kg-NO ₂ yr^{-1}	6.27 e-6	0.71	0.231
HCl		kg-HCl yr^{-1}	2.28 e-5	2.60	0.842
NH ₃		kg-NH ₃ yr^{-1}	5.24 e-5	5.97	1.93

Type 2: Emission unit is in kg of S, N, or Cl yr^{-1}

Causative substance		Emission unit	Midpoint approach		Endpoint approach
			ADF ^a	AP ^b	DF ^c
SO ₂		kg-S yr^{-1}	1.75 e-5	1.00	0.646
NO _x	NO	kg-N yr^{-1}	2.06 e-5	1.18	0.729
	NO ₂				
HCl		kg-Cl yr^{-1}	2.35 e-5	1.34	0.866
NH ₃		kg-N yr^{-1}	6.36 e-5	3.63	2.34

^a ADF (Atmospheric Deposition Factor): Utilizable for Category Indicator (CI) with emission inventory of substance j , $inv(j)$ [kg]. $CI = \sum inv(j) \times ADF(j)$. CI has a unit of $\text{eq km}^{-2} \text{yr}^{-1}$ which indicates a mean increase of H^+ deposition load due to additional emissions of causative substances.

^b AP (Acidification Potential): Equal to SO₂ equivalent ADF.

^c DF (Damage Factor): Utilizable for Damage Indicator (DI) with $inv(j)$ [kg]. $DI = \sum inv(j) \times DF(j)$. $DF(j) = ADF(j) \times EF$. DI has a unit of kg yr^{-1} which indicates the total NPP loss in Japan due to additional emissions of causative substances.

2.4 Category indicator

CI indicates the total increase of H^+ load to the surface ($\text{eq km}^{-2} \text{yr}^{-1}$) corresponding to an emission inventory. CI is given by,

$$CI = \sum_j^{SO_2, NO, NO_2, HCl, NH_3} Inv(j) \cdot ADF(j) \quad (12)$$

where $Inv(j)$ denotes an emission of causative substance, j (kg yr^{-1}).

2.5 Damage indicator

DI indicates the total NPP damage in all of Japan (kg-NPP yr^{-1}) corresponding to an emission inventory. DI is given by,

$$DI = \sum_j^{SO_2, NO, NO_2, HCl, NH_3} Inv(j) \cdot DF(j) \quad (13)$$

3 Discussion

3.1 Category demarcation

There are some difficulties in clearly demarcating impact categories regarding air pollutants but excluding chemicals, *i.e.* acidification, eutrophication, photochemical oxidant, and regional air pollution, since causative substances and endpoints of these categories are common and/or interactive. However, relevant processes of each impact category are largely different both in pathways and the seriousness of their impact; it forces a demarcation of impact categories regarding air pollution.

The consensus of handling these categories in LIME is as follows: direct effects of air pollutants were divided into photochemical oxidant and regional air pollution considering strong acute toxicity of oxidants and chronic toxicity of other air pollutants, and then indirect effects of air pollutants were involved in acidification. Eutrophication through the atmospheric deposition was also a subject of acidification; eutrophication of closed water body was independently categorized (see Fig. 1). However, eutrophication caused by atmospheric deposition was excluded from quantification of damage at the present due to the difficulty in evaluation of damage, since eutrophication has aspects of both merit and demerit to ecosystems.

3.2 Cause of impact and impact indicator

Aluminum toxicity is well known as one of the hypotheses of forest decline (Hinrichsen 1986) that was selected in this study as the main cause of indirect effects on terrestrial ecosystems induced by acidification. However, there is room for considering other causes of impact regarding indirect effects of soil acidification such as base cation deficiency and heavy metal toxicity.

The concentration of leachate Al^{3+} without threshold value, therefore being essentially toxic, was applied to express the aluminum toxicity in this study. Another index, e.g. the BC/Al ratio (Sverdrup and Warfvinge 1993), the molar ratio of base cations ($\text{K}+\text{Mg}+\text{Ca}$) to aluminum with threshold value, is ponderable for consideration in a future study. At the present, however, no spatial information on leachate concentration of base cation was available in all of Japan. It should be noted that only one tree species was involved to express the relationship of aluminum toxicity to plant growth, which is a weak point in the method. This is a subject to be improved. A discussion of the representativeness of this relational expression of one tree species is to be seen in section 3.4.

NPP was selected as the impact indicator of terrestrial ecosystems in this study. As shown in Fig. 1, there are two safeguard subjects on ecosystems in the framework of LIME, i.e. primary production and biodiversity. NPP is an indicator expressing primary production. As mentioned in the introduction, biodiversity was excluded from the impact category of acidification, regretfully due mainly to a lack of information. Acidification may also possibly affect biodiversity through its effects on interspecies competition, due to the differences in sensitivity against acidic condition. The perturbation of interspecies competition, however, is difficult to model. Because insufficient information is available for all aspects, not only acid tolerance, but also competition mechanism among various species, and the chain or consequence of the change triggered by the first impact, it is very difficult to quantitatively evaluate the change in biodiversity quality, since it does not always entail extinction.

We should discuss why the buffering capacity of soil to neutralize acid was excluded in this study. This property is never avoidable when dealing with inland water impact due to acidification, since inland water is what exudes through its watershed, i.e. soil and surface geology. In general, the total of these buffering capacities are large enough to neutralize substantial acid. This means that the low level of acid load which we consider in LCIA may bring about a negligible

effect. Regarding terrestrial ecosystems, the impact of aluminium toxicity on plant takes place on the root system hindering such features as root elongation and nutrient uptake (Cronan 1994). Fine roots, taking on the primary role of nutrient uptake, are mainly distributed in the surface layer of soil. At the surface layer, atmospherically loaded acid can affect plant roots before being neutralized. Because the surface layer is also the boundary between air and soil, and the input acid then has a chance to induce effects rapidly. Still, it is natural that some of the acid is neutralized even at the surface layer. However, it was difficult to mechanistically determine how much of this acid will be neutralized. Thus, the buffering capacity of soil to acid was excluded in this study. In this sense, our estimation is related to the side of safety.

3.3 Geographical boundary

This study limited the geographic boundary of acidification within Japan to both the emission of causative substances and the impact on terrestrial ecosystems. In fact, acidification frequently becomes a transboundary issue typically emerged in Europe. Japan is an emitter of causative substances of acidification and simultaneously a receptor of acidifying substances in East Asia. It is no doubt that atmospheric deposition in Japan is also effected by the pollutants from surrounding countries, e.g. China and South Korea. This could be involved in the baseline of acidic deposition if observation data were applied to determine this baseline. In this study, instead of directly including the baseline to estimate EF, however, the initial soil pH was assumed to be at equilibrium to the baseline of acidic deposition. Meanwhile, the aim of LIME was to evaluate additional effects caused by some alternatives in Japanese industries. Hence, the first purpose of this study was to evaluate effects due to additional acid loads within Japan. In fact, however, there are many Japanese companies that have moved their production bases to abroad such as China. Thus, the transboundary, source-receptor relationships among countries in East Asia should be involved in the method in the future.

3.4 Parameter representativeness

SRR directly and proportionally affects the damage function of acidification so that it is important to know the fluctuation range of SRR for each causative substance, which changes year by year affected by conditions of meteorology and emission. The SRRs determined for respective causative substances in this study were fixed values due to limited data availability; its representativeness and uncertainty are subjects to be examined. The SRRs of HCl and NH_3 were three-fold larger than that of SO_2 and NO_x . The estimation method applied to determine the SRRs of HCl and NH_3 was relatively rough. Thus, the uncertainties of their values are larger than that of SO_2 and NO_x , which brings about difficulties in comparison. However, the difference can be attributed, in part, to the properties of HCl and NH_3 , i.e. highly water soluble and then high velocities of dry deposition (Puxbaum and Gregori 1998). Although SO_4^{2-} and NO_3^- generated from oxidizing SO_2 and NO_x are also highly water soluble, oxidization needs time to progress. Therefore, SO_2 and NO_x tend to be transported long-range during this time, which results in smaller SRRs.

Although Japanese red pine is one of the common tree species in Japan, it is rather a pioneer species and its forests cover small areas in Japan. However, no similar model to Eq. 8 was derived regarding other tree species that consist of dominant vegetation in Japan. Accordingly, regardless of vegetation legends, Eq. 8 was applied to express the relationship between leachate Al^{3+} concentration and the growth rate of a tree. The sensitivity of Japanese red pine to aluminum tended to be higher than that of *Fagus crenata* (Japanese beech) from experiments on the BC/Al ratio (Izuta *et al.* 2001). Thus, it is possible to overestimate NPP damage in Eq. 8.

The correction using NDVI for NPP values (see 1.5.4) was to describe the difference of NPP by climate and landform, since the original NPP dataset (Iwaki 1981) only gives fixed values. This correction method was developed and discussed in Nakagawa *et al.* (2002) as a part of LIME. The result could reflect the changes in NPP by climate, though it perhaps has some uncertainty.

3.5 Results of damage function

The DFs obtained in this study showed the similar order of values compared to emission of causative substances on mass basis (see Table 4). It was just a coincidence coming down to the size of geographical boundary, limited to Japan. Namely, a value of EF becomes large if the range of geographical boundary was extended; it directly results in an increase of land area where its NPP damage should be summed. On the other hand, a value of ADF is relatively stable regardless of the size of geographical boundary, unless a value of SRR changes remarkably.

The relationship between increased H^+ deposition and total NPP damage in Japan, i.e. EF, showed complete linearity (see Fig. 3). However, even the uppermost value of increased H^+ deposition, $200 \text{ eq km}^{-2} \text{ yr}^{-1}$, accounts for only 0.2% of the baseline H^+ load in Japan, around $100,000 \text{ eq km}^{-2} \text{ yr}^{-1}$ ($1000 \text{ eq ha}^{-1} \text{ yr}^{-1}$). Namely, the range of increased H^+ deposition is within very low loads and the relationship could keep its linearity, although non-linear relationships were used for determining the EF such as H^+ concentration and pH, pH and aluminum concentration, and aluminum concentration and plant growth. It is, however, important to notice that the low H^+ load, $200 \text{ eq km}^{-2} \text{ yr}^{-1}$, corresponds to 74 Meq yr^{-1} of additional H^+ load in the whole of Japan. It is equivalent to 3.5 Gg of SO_4^{2-} or 4.6 Gg of NO_3^- in charge balance, all together no small amount. The usage of the method is mainly to estimate additional impact due to some alternation in industry. Accordingly, the range of increased H^+ deposition seems to be sufficient. More attention is also needed to evaluate the magnitude of total NPP damage. As shown in Fig. 3, the order of total NPP damage is 10^6 kg yr^{-1} , which indicates the nationwide sum of NPP damage in Japan. This damage is equivalent to $3 \text{ kg km}^{-2} \text{ yr}^{-1}$ ($0.03 \text{ kg ha}^{-1} \text{ yr}^{-1}$), considering the area of Japan. On the other hand, NPP itself is on the order of $10,000 \text{ kg ha}^{-1} \text{ yr}^{-1}$. Consequently, the NPP damage which is involved in this study contributes to one millionth of the NPP itself, which sounds like quite a risk.

NH_3 showed the largest DF and HCl was next, although SO_2 and NO_x are major causative substances of acidification. The result was ascribed to the relatively large SRR and low mo-

lecular weight for NH_3 and the relatively large SRR for HCl (see Table 3). The result is likely to include the fact, but the uncertainties of DFs for NH_3 and HCl are essentially larger than that of SO_2 and NO_x . A lot of attention is needed to handle the difference of DFs, and the reliability of relevant parameters, SRR in particular, should be enhanced. In addition, SO_2 and NO_x perhaps dominate emission inventory in industry, since these substances are evidently generated in quantity by energy consumption compared to HCl and NH_3 .

3.6 Comparison with another study

As shown in Table 1, it was difficult to compare our damage function directly to other endpoint approaches because of the differences in frameworks. Here, a comparison on the midpoint level between our result and the result of Heijungs *et al.* (1992) updated with Hauschild and Wenzel (1998) was carried out. The value of AP in this study for each causative substance, $AP(j)$, was derived as follows:

$$AP(j) = ADF(j) / ADF(\text{SO}_2) \quad (14)$$

Table 5 shows the comparison result. APs of SO_2 and NO_x were similar in both study results. APs of HCl and NH_3 , on the other hand, were different and our result showed larger values. The reason was ascribed to the SRR. The similar value of SRR for NO_x to that of SO_2 (see Table 3) brought about the similar APs between both study results, although Heijungs *et al.* (1992) took no consideration of SRR. However, the larger values of SRR of HCl and NH_3 than that of SO_2 (see Table 3) resulted in the larger values of AP (Table 5) in this study. The SRR is indispensable to determine AP, though it perhaps has considerable uncertainty. Simply because acidification impact takes place on the surface and not in the atmosphere. Accordingly, an improvement of the reliability of SRR is important.

Table 5: Comparison of Acidification Potential (AP)

Causative substance		AP ^a	
		This study	Heijungs <i>et al.</i> (1992)
SO_2		1.00	1.00
NO_x	NO	1.09	1.07
	NO_2	0.71	0.70
HCl		2.60	0.88
NH_3		5.97	1.88

^a AP: kg-SO_2 equivalent kg^{-1} .

^b The result of Heijungs *et al.* (1992): updated with Hauschild and Wenzel (1998).

4 Conclusion and Future Work

The damage function of acidification for terrestrial ecosystems easily gives concrete NPP damage due to an additional emission of its causative substance. The impact indicator, NPP, also has an advantage in its unit, mass, that is directly summable through the entire impact categories, and provides a chance of comparison between other impact indices such as crop production and fishery production.

Expansion of endpoints, not only NPP damage of terrestrial ecosystems but also reproduction damage of aquatic ecosystems, material degradation, human health with regard to

drinking water toxicity, and of course the aspects of biodiversity, is an important subject for future work to comprehend the present LCIA framework of the impact category of acidification. Further, uncertain analyses for major relevant parameters will provide helpful information on the reliability of damage function. The SRR, in particular, plays an important role in determining the effects of a load of acid on the surface through the atmosphere.

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